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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of speciated and total mercury dry deposition at monitoring locations in Eastern and Central North America" *by* L. Zhang et al.

Anonymous Referee #1

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This manuscript addresses some of the shortcomings of previous inferred dry deposition studies by comparing their dry deposition estimates to measurements of Hg in environmental media. The results presented in this manuscript are important.

The limitations of GOM measurements using KCl denuders is not sufficiently discussed. In particulare there needs to be a discussion of GOM measurement artifacts when using KCl denuders in the presence of oxidants and how that uncertainty may impact the results (see Lyman et al. 2010b, it is cited in the manuscript but not discussed).

In several areas the manuscript describes results as "significant", describes results as

"better" and quantifies errors and uncertainty but the statistical metrics used to assess these results are not given. In general this manuscript would benefit from a better quantification of the results and the use appropriate statistical metrics.

There are some typos and some of the language used in this manuscript is incorrect and the manuscript could benefit from a thorough proof reading.

The magnitude of the GEM re-emissions and dry deposition velocity appear to be critical in determining the net GEM flux and the magnitude of the total Hg deposition. The uncertainty in the total Hg deposition to these variables should be discussed more given the uncertainties in them.

The comparison with throughfall measurements is a strong point in this manuscript. However, I would like to see a more quantitative presentation and discussion of these results, e.g. plotting throughfall deposition totals, and modeled totals as a function of the GEM, GOM, PBM concentrations or plotting the throughfall deposition totals as a function of the number of Hg sources in a 100 km radius. If GEM dry deposition is driving the total Hg dry deposition to these ecosystems, then the throughfall measurements should be better correlated with the ambient GEM concentrations than GOM or PBM.

Specific comments:

Page 2791 Line 5: What exactly are the input parameters scaled by LAI?

Page 2792 Lines 5-8: Is there a means of placing a bounds on the error that soil emissions may cause in net GEM flux, e.g. a model sensitivity?

Page 2792 Lines 20-21: Six sites have both dry deposition and litterfall measurements. Why not look at all the litterfall sites. If GEM is the predominant deposited species, we would expect relatively uniform litterfall deposition values because GEM has a relatively long atmospheric lifetime/or is constantly recycled resulting in relatively uniform concentration fields.

Section 3.1: This section could generally benefit from a more quantitative discussion.

Page 2794 Lines 2-3: "GOM and PBM where highly variable ... much larger than GEM" The authors should quantify this?

Page 2794 Lines 3-4: "concentrations in spring were much higher" How much higher?

Page 2794 Lines 5-7: "At two other urban/suburban sites..." Which sites?

Page 2794 Line 8: "For PBM, the highest season concentrations..." This was also observed by Amos et al 2012 and they derived an empirical PBM - GOM partitioning model to describe this variability. Page 2794 Line 15: "dry deposition theory" I have heard of that and if the authors are going to stick with it they need to provide a citation. The following discussion appears to discuss the precipices of boundary layer meteorology and similarity theory as they relate to air-surface exchange (see Stull 1988). Alternatively the authors could cite micormeteorological theory (see Baldocchi et al 1988).

Page 2795 Lines 2-3: "turbulence intensity (friction velocity)" The friction velocity, u*, is not turbulence intensity and is not defined in the manuscript or alluded to why it would be important. Mechanical turbulence intensity, what the authors are discussing, is oftern defined as the standard deviation of the mean wind speed divided by the mean wind speed. u* is the shear stress defined as the square root of magnitude of the surface Reynold's stress divided by the atmospheric density and is an important variable in deriving the aerodynamic and boundary layer resistances used in estimating dry deposition.

Page 2796 Lines 1-2: "significant Hg emissions" What makes these Hg emissions significant?

Page 2796 Lines 6-8: Is this an artifact of estimating the new GEM flux and are there litterfall measurements to support this claim?

Page 2796 Line 29: "Good agreement between ..." What was used as a measure of

good? How was this quantified. Because these estimates where within the range of surrogate surface measurements?

Page 2797 Line 15: "Great Lakes region" This is also qualitatively shown for CMAQ and CAMx at AMNet sites in Baker and Bash 2012.

Section 3.4: The impact that oxidants have on GOM measurements using KCl denuders should be discussed in this section (see Lyman et al. 2010b).

Page 2798 Line 11: "lower than 40%" How was this determined?

Page 2798 Line 20: "should be generally within a factor of 2" Why would that be? If GOM deposits like HNO3 then the uncertainty would be dominated by uncertainties in the aerodynamic boundary layer resistances because canopy resistances are believed to be near zero (see Flechard et al. 2011). It is unclear how that equates to a factor of 2.

Page 2798 Line 23: "within 30% differences" If this is just at several sites, which sites were included in the comparison and how does it compare to all the collocated sites.

Page 2799 Line 2: "only slightly" Is this referring to the 30% difference mentioned on the previous page? If not, please quantify this statement.

Page 2799 Lines 27-29: Doubling GOM+PBM deposition may not change the conclusion but changing your GEM dry deposition due to inaccurate emission estimates could. The net GEM flux should be discussed in more detail.

Page 2800 Lines 23-24: How was significance determined?

Page 2801 Line 12: "with low soil Hg emissions" is unsupported by the cited literature or research presented in the manuscript. Graydon et al. 2009 used isotropically labeled Hg to show that the foliar Hg did not contain a large contribution from the soils at ELA.

Page 2802 Lines 1-2: "are at best model estimates with large uncertainties" Please quantify what large uncertainties means.

Technical corrections

Page 2794 Line 15: "simialr" should be "similar"

References

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